BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images may include (but are not limited to):

- BLACK BORDERS
- TEXT CUT OFF AT TOP, BOTTOM OR SIDES
- FADED TEXT
- ILLEGIBLE TEXT
- SKEWED/SLANTED IMAGES
- COLORED PHOTOS
- BLACK OR VERY BLACK AND WHITE DARK PHOTOS
- GRAY SCALE DOCUMENTS

IMAGES ARE BEST AVAILABLE COPY.

As rescanning documents *will not* correct images, please do not report the images to the Image Problem Mailbox.

(12) UK Patent Application (19) GB (11) 2 188 559 (13) A

(43) Application published 7 Oct 1987

(21) Application No 8705753

(22) Date of filing 11 Mar 1987

(30) Priority data

(31) 3608635 (32) 14 Mar 1986 (33) DE

(71) Applicant
Drache Keramikfilter Produktions-GmbH,

(Incorporated in FR Germany),

Werner-von-Siemens-Strasse 9, 6252 Diez, Federal Republic of Germany

(72) Inventors
Helmut Schweers Dipl.-Ing,
Günther Förster Dipl.-Ing,
Frank Drache Dipl.-Betriebswirt

(74) Agent and/or Address for Service
F. J. Cleveland & Company, 40-43 Chancery Lane, London WC2A 1JQ

(51) INT CL⁴
B01D 53/36

(52) Domestic classification (Edition I)
B1F D2D
U1S 1851 B1F

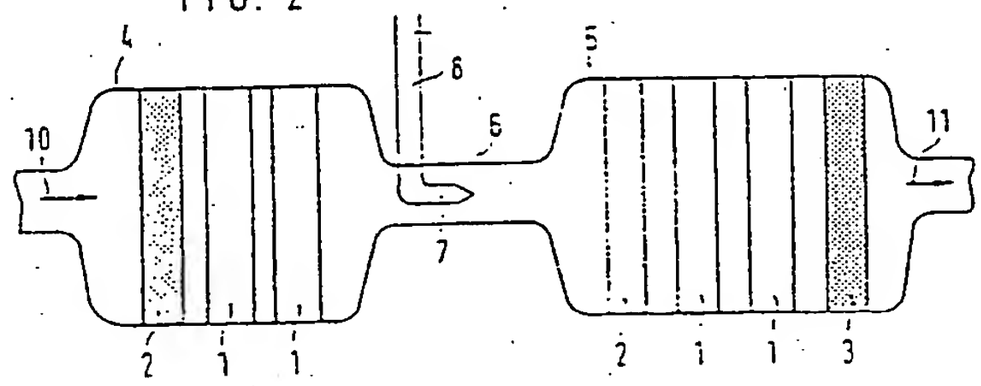
(56) Documents cited
GB 1461804 US 4581206
GB 1455433 US 4425304
GB 1406704 US 4278639

(58) Field of search
B1F
Selected US specifications from IPC sub-class B01D

(54) Exhaust gas reactor

(57) An exhaust gas reactor comprises at least first and second open-pore ceramic foam plates which are arranged in succession in the direction of flow through the reactor. The plates of ceramic foam have a catalytically active surface layer thereon. Filter plates of uncoated ceramic foam may additionally be provided upstream and downstream of the first-mentioned plates.

FIG. 2



GB 2 188 559 A

FIG. 1

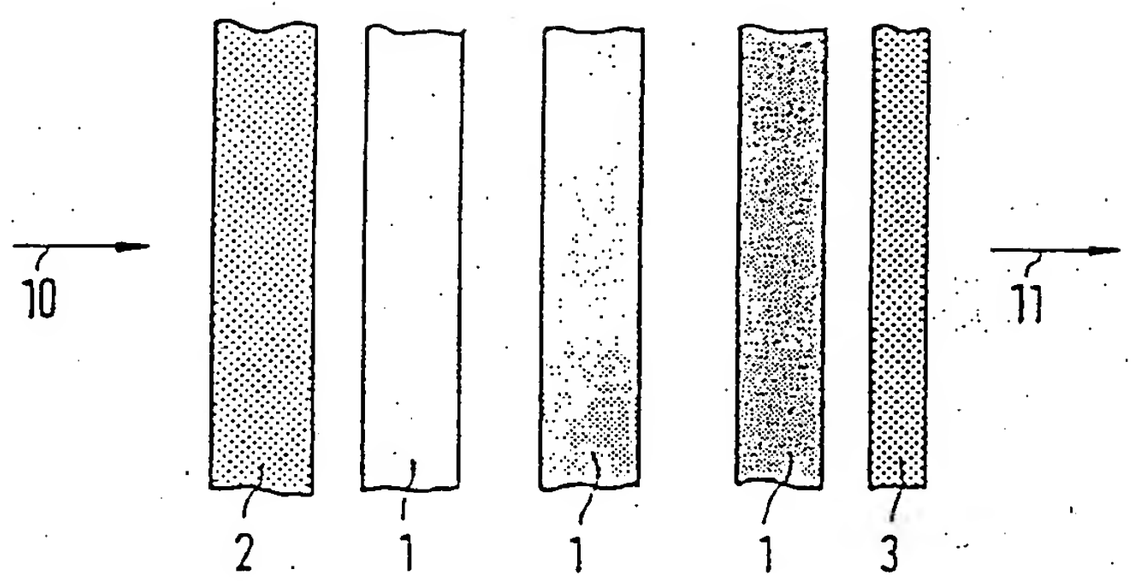
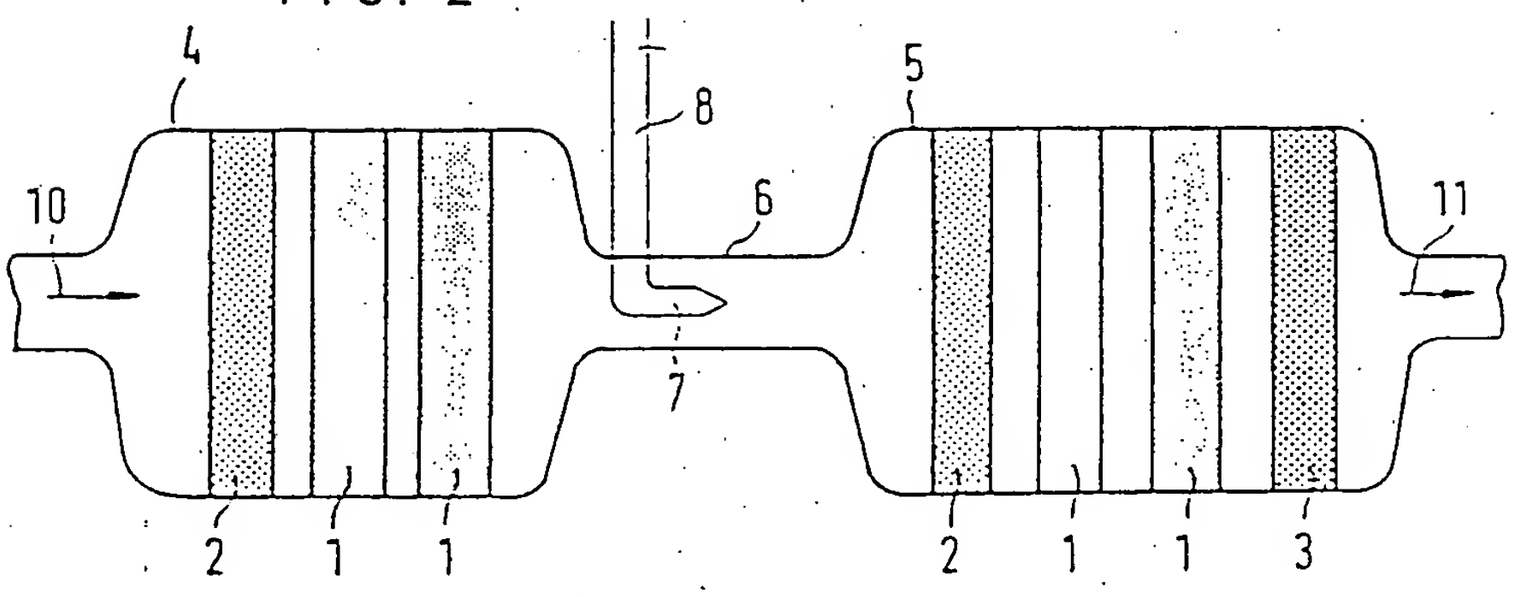


FIG. 2



SPECIFICATION

Exhaust gas reactor and process for the production thereof

The invention relates to an exhaust gas reactor, for example for use with an internal combustion engine, and a process for the production of such a reactor.

The term exhaust gas is used in this specification in a broad sense and may therefore embrace exhaust gas from an internal combustion engine as well as exhaust or waste gas from other sources, such as a paint spraying booth.

One form of an exhaust gas reactor comprises a refractory carrier material in the form of a monolithic block of open pore ceramic foam, which has thereon a catalytically active surface layer.

Porous ceramic foam is produced by an open-pore plastics foam being filled with a slurry of ceramic material, and then vaporising the plastics foam at elevated temperature. The porous ceramic foam produced in that way is also of an open-pore nature and is then provided with a catalytic layer which is matched to the respective chemical process involved. Catalysts produced in that way from porous ceramic foam are subjected to further processing to produce monolithic blocks of appropriate size, and used in that form.

According to the present invention, there is provided an exhaust gas reactor including a refractory carrier means of open-pore ceramic foam comprising at least two plates which are disposed in succession in the direction of gas flow through the reactor, and a catalytically active surface layer on the carrier means.

Tests have been carried out which show that an exhaust gas reactor which is constructed from a plurality of successively disposed plates is substantially more efficient in operation than an exhaust gas reactor consisting of a monolithic block of appropriate size. Furthermore, an exhaust gas reactor comprising a plurality of plates is substantially easier to manufacture and it is also easier to provide it with a catalytically active surface layer, than a large monolithic block. It is possible to arrive at a made-to-measure construction, depending on the particular composition and temperature of the waste or exhaust gases which are to be subjected to the post-combustion procedure, by virtue of the plates being of suitable composition, configuration and dimensions. In addition, the reactor can be rapidly adapted to the respective operating conditions involved, particularly in the event of changes therein, by virtue of the insertion or replacement of plates. All in all, because of the large internal surface area of the ceramic foam, the exhaust gas reactor can be small in volume, in relation to the respective gas through-put thereof. A compact exhaust gas reactor of that nature is particularly important when, in connection with subsequent fitting to motor vehicles, a conventional silencer box or muffler is to be replaced by an exhaust gas reactor which must take account of the predetermined dimensions and conditions of installation of the respective silencer box or muffler.

Besides the very large internal surface area of a ceramic foam, relative to its external volume, a critical consideration in regard to the level of efficiency of an exhaust gas reactor in accordance with the present invention is also the high level of reaction probability which is provided by the micro-turbulent post-flow of the individual rods or fibres of the ceramic foam.

In accordance with a further feature of the invention, the porosity of the plates is between 10 and 60 ppi (pores per inch), and preferably between 10 and 30 ppi. The reference to pores per inch (ppi) denotes the number of pores per inch as a unit of length.

A further embodiment of a reactor according to the invention advantageously provides that plates of a different configuration or nature may be used. The different configuration or nature may be that the porosity of the plates is different, for example a plate with coarse pores is followed by one or more plates with finer pores. However the different configuration or nature may additionally or alternatively involve different catalytically active surface layers. Furthermore and possibly in addition thereto the plates may be of different shapes and sizes in order for example to produce a given flow characteristic in aerodynamic respects.

A further feature of the invention may provide that one or more uncoated plates is or are arranged upstream and/or between and/or downstream of catalytically coated plates. In that connection the uncoated plates act in particular as a filter means. In that way, undesirable particles in the gas flowing into the reactor are filtered out of the gas flow prior to passing into the part of the reactor in which reaction occurs, and reaction products are filtered out of the gas flow if further uncoated plates are arranged within the coated plates of the reactor or downstream thereof. The pore size of the uncoated filter plates is to be matched to the size of the particles which are to be filtered out of the gas flow. If an uncoated plate with very fine pores is disposed at the downstream end of the reactor assembly, it is possible to ensure that any particles of the ceramic carrier material which come loose do not pass into the atmosphere, together with constituents of the washcoat base layer and the catalyst material.

In one embodiment of a reactor, the individual plates thereof may bear against each other in a sandwich-like construction. An advantageous feature of the invention provides however that the plates are arranged at a spacing from each other, in which respect the spacings between the plates may be the same or may vary. The spacings between the plates prevent the gases from forming preferential through-flow paths, which would have the result that parts of the plates would not have gas flowing therethrough or would have less gas flowing therethrough, so that the level of efficiency would drop, in relation to unit of surface area. If the spacing between plates is correctly chosen, turbulent effects occur between the plates, which ensure that the gas flow is balanced and equalised out. The spacings between the plates are also to be selected having regard to the flow speeds through the reactor, the

dimensions of the plates and the properties of the respective gases involved. In that connection however the spacings should be kept as small as possible, having regard to the reactor being of a compact design.

If, in accordance with a further advantageous feature of the invention, the plates are individually interchangeable, then a reactor of that nature is simpler and less expensive from the point of view of maintenance than a reactor which uses a monolithic block. In that case individual plates can be replaced for repair or exchanged for other plates, if the operating conditions alter. To remove the dust which has been produced, the above-mentioned uncoated filter plates may be exchanged or only cleaned after they have been removed.

A particularly advantageous development which is also an aspect of independent significance provides that the catalyst material used comprises at least one non-noble metal oxide.

The reaction temperature of such oxides is higher than that of noble metals which are conventionally used, for example platinum, and is between about 450 and 750° C. As a result, the level of effectiveness scarcely decreases, even under high loading and at high temperatures, so that such a reactor is particularly suitable not just for internal combustion engines of motor vehicles but also for stationary engines. The non-noble metal oxides are also resistant to lead poisoning phenomena and to other noxious substances such as chlorine, fluorine and sulphur, as occur in the uncleaned gases of stationary engines.

In a preferred feature of the invention, the catalytically active surface layer of a non-noble metal oxide may be arranged on what is known as a washcoat base layer of aluminium oxide in its γ-modification form. Such a washcoat base layer is known and provides for a more rapid and better reaction. In order to improve long-term stability, additives may be added to the base layer, for example compounds of elements from the rare earth group.

In order to provide a reducing reaction, a further embodiment of the invention provides for using a catalyst material comprising at least one oxide of the following metals: iron (Fe_2O_3 or Fe_3O_4), cobalt (CoO or Co_2O_3 or Co_3O_4), nickel (NiO or Ni_2O_3), titanium (TiO_2), vanadium (V_2O_5), chromium (Cr_2O_3), molybdenum (MoO_3), tungsten (WO_3), and manganese (MnO or Mn_2O_3 or Mn_3O_4), or at least a mixture of at least two thereof.

To provide an oxidising reaction, the catalyst material of the reactor may comprise at least one oxide of at least one of the following metals: copper (Cu_2O), chromium (Cr_2O_3), molybdenum (MoO_3), tungsten (WO_3), manganese (MnO or Mn_2O_3 or Mn_3O_4), vanadium (V_2O_5), iron (Fe_2O_3) or at least one mixture of at least two thereof.

In a further advantageous embodiment of the invention, the reactor may be of a two-stage configuration, with a reducing reaction taking place in the first stage and an oxidising reaction taking place in the second stage. For example, in the first stage of an exhaust gas reactor for internal combustion engines, nitrogen oxides (NO_x) are reduced to harmless nitrogen at the catalyst material by means of the carbon monoxide (CO) in the exhaust gas. Then, the second stage involves oxidation of the excess carbon monoxide to form carbon dioxide, with oxygen possibly also being added, in the form of air. In other areas of use of such a reactor, it is also possible to provide for the oxidation of other constituents, in particular organic components, for example hydrocarbons. Such constituents occur inter alia in the waste or exhaust gases from paint spraying booths. The two stages of the reactor each have at least one plate and may be arranged in separate housings or in a common housing. At least one uncoated filter plate is desirably arranged upstream of the first stage, so such a filter can also retain soot particles which can then undergo post-combustion if the initial filter reaches a temperature above 600° C. The second stage may be followed by an uncoated fine-pore filter plate, constituting a very fine filter which retains in particular pieces of ceramic carrier material which have come away.

In another aspect, the present invention also provides a process for the production of an exhaust gas reactor, wherein a non-noble metal oxide is added to the slurry which, as referred to in the opening part of this specification, is used to produce the ceramic foam from open-pore plastics foam. After calcination of the foam which is coated with the ceramic material, the non-noble metal oxide is then a constituent of the ceramic material and is thus also present at the surface thereof. Such a mixing effect may be used in particular when the oxide is comparatively inexpensive.

An alternative option in regard to the process for producing the exhaust gas reactor according to the invention provides that the foam which has been coated with ceramic material and pre-dried but not yet subjected to the calcination operation may be coated, preferably by a dipping operation, with a particular suspension or slurry which contains the non-noble metal oxide and optionally further constituents which promote adhesion of the oxide as well as the catalytic function. Besides for example titanium dioxide, the suspension or slurry may additionally contain the material for the base layer (aluminium oxide).

Another form of procedure for applying the catalyst material in the reactor according to the invention provides that the foam which has been coated with ceramic material and possibly the base layer and which may or may not have been subjected to the calcination operation is impregnated with a solution of a metal salt of the non-noble metal to be used, and then the metal is converted into the metal oxide by a heat treatment such as calcination, and fixed on the ceramic foam.

The reactor according to the invention is inexpensive to manufacture, enjoys a wide range of adaptability and has a high level of efficiency, even at high temperatures, while being easy and simple to maintain and being capable of reacting to altering operating conditions.

Embodiments of a reactor according to the

invention and the process for the production thereof will now be described by way of example with reference to the accompanying drawing in which:

Figure 1 is a diagrammatic view in longitudinal section of a first embodiment of the reactor according to the invention, and

Figure 2 is a basic view in longitudinal section of a two-stage reactor as a second embodiment of the invention.

Referring to the diagrammatic view in Figure 1, an exhaust or waste gas reactor according to the invention comprises three plates 1 of open-pore ceramic foam with a catalytically active surface layer thereon, which are arranged in succession in the direction in which the gas flows through the reactor, as indicated by the arrows 10 and 11.

Disposed upstream of the first reactor plate 1 is a further plate 2 of open-pore ceramic foam, which is not coated with catalyst material. The plate 2 serves as a filter plate for particles contained in the flow of exhaust gas. Furthermore, arranged downstream of the last reactor plate 1 is a very fine filter plate 3 of open-pore ceramic foam without a catalyst coating thereon. The plate 3 has very fine pores and filters out of the gas flow particles of the ceramic base material which have come loose. The spacings between the individual plates 1, 2 and 3 provide for a turbulent effect therein. These spacings are selected to be non-uniform in such a way that the reaction is as complete as possible, the flow resistance is at a low level and the dimensions of the arrangement are not excessive.

Figure 2 is a diagrammatic view of a two-stage reactor. The two stages are arranged in separate casings or chambers 4 and 5 which are in communication by way of a pipe 6. The waste or exhaust gas to be treated flows into the casing 4 in the direction indicated by the arrow 10. Arranged first in the casing 4 is an uncoated filter plate 2 which is followed at a spacing by two spaced-apart reactor plates 1 which are coated with catalytically active material. In the plates 1, the nitrogen oxides (NO_x) in the gas react with the carbon monoxide (CO) which is also present therein, with the nitrogen oxides being reduced to non-toxic nitrogen. In the pipe 6, air is then added to the exhaust gas by means of a nozzle 7 from a pipe 8, for example by means of a pump in the form of a blower. If it is assumed that the denoxed gas in the pipe 6 still has a component of about 1% CO, then about 0.5% O_2 from the air must be added for complete combustion. With an oxygen component for about 20%, that means the addition of about 2.5% air. As operation is usually conducted at higher than stoichiometry, about double the amount is added in a practical situation, that is to say about 5% air.

In the casing 5, the gas again first flows through a filter 2 and then two reactor plates 1, at the surface of which the carbon monoxide undergoes combustion to form carbon dioxide. A very fine filter plate 2 at the outlet of the casing 5 eliminates reaction particles and fine dusts.

One embodiment of the reactor according to the invention used chromium trioxide as the metal oxide. The ceramic foam carrier material was impregnated with ammonium dichromate solution (between 3 and 25%) and then the substance was thermally decomposed to form chromium trioxide (Cr_2O_3), on the carrier material. The ammonium dichromate may also be reduced with a reducing agent such as for example ethanol or ascorbic acid, to form trivalent chromium trioxide. Fixing on the carrier material is then effected by drying and calcining. The thermal expenditure involved in such a procedure is slight.

Various forms of process may be employed for producing a reactor as described above; for example, when using a slurry of ceramic material to fill an open-pore plastics foam which is then vaporised at elevated temperature to produce a porous ceramic foam, the catalyst material such as the non-noble metal oxide referred to above may be added to the slurry used to fill the plastics foam which is then removed in the manner stated. Alternatively, the foam may be coated with ceramic material and then impregnated with a solution of a salt of a non-noble metal, the salt then being converted into the metal oxide and fixed on the ceramic foam.

It will be appreciated that the above-described embodiments of the reactor and process according to the invention have been set forth solely by way of example thereof and that various modifications and alterations may be made therein without thereby departing from the scope of the invention as defined by the appended claims.

CLAIMS

1. An exhaust gas reactor including a refractory carrier means of open-pore ceramic foam, comprising at least two plates which are disposed in succession in the direction of gas flow through the reactor, and a catalytically active surface layer on the carrier means.

2. A reactor according to claim 1 wherein the porosity of the plates is between 10 and 60 ppi.

3. A reactor according to claim 2 wherein the porosity of the plates is between 10 and 30 ppi.

4. A reactor according to claim 1 and comprising plates of a different configuration or nature.

5. A reactor according to claim 4 wherein the porosity of the plates is of different sizes.

6. A reactor according to claim 4 or claim 5 wherein the plates have different catalytically active surface layers.

7. A reactor according to any one of claims 4, 5 and 6 wherein the plates are of different shapes.

8. A reactor according to any one of the preceding claims and comprising one or more uncoated plates disposed upstream and/or between and/or downstream of the catalytically coated plates.

9. A reactor according to one of the preceding claims wherein the plates are arranged at a spacing from each other.

10. A reactor according to claim 9 wherein the spacing between the plates varies.

11. A reactor according to one of the preceding claims wherein said plates are individually interchangeable.

12. A reactor according to one of the preceding claims wherein the catalyst material used includes at least one non-noble metal oxide.

13. A reactor according to one of the preceding claims wherein said catalytically active surface layer is arranged on a base layer (washcoat) of aluminium oxide (Al_2O_3) in the γ-modification form.

14. A reactor according to claim 12 or 13 wherein, for a reducing reaction, the catalyst material comprises at least one oxide of at least one of the following metals: iron, cobalt, nickel, titanium, vanadium, chromium, molybdenum, tungsten, and manganese or a mixture of at least two thereof.

15. A reactor according to claim 12 or claim 13 wherein, for an oxidising reaction, the catalyst material comprises at least one oxide of at least one of the following metals; copper, chromium, molybdenum, tungsten, manganese, vanadium, iron or a mixture of at least two thereof.

16. A reactor according to one of the preceding claims which comprises at least first and second stages, of such a nature that in operation a reducing reaction takes place in the first stage and an oxidising reaction takes plate in the second stage.

17. A reactor according to claim 16 wherein said first and second stages each comprise at least one plate and they are arranged in a common housing.

18. A reactor according to claim 16 or claim 17 and further including an uncoated filter plate arranged upstream of the first stage in the direction of flow therethrough.

19. A reactor according to one of claims 16 to 18 and further comprising an uncoated fine-pore filter plate arranged downstream of the second stage in the direction of flow therethrough.

20. A reactor according to one of claims 16 to 19 wherein air is injected into the flow of gas through the reactor, between said first and second stages.

21. An exhaust gas reactor substantially as hereinbefore described with reference to Figure 1 of the accompanying drawings.

22. An exhaust gas reactor substantially as hereinbefore described with reference to Figure 2 of the accompanying drawings.

23. A process for the production of a reactor according to any one of claims 12 to 20 comprising introducing a slurry of ceramic material into an open-pore plastics foam and then removing the plastics foam, wherein said non-noble metal oxide is added to the slurry used to produce the ceramic foam from the open-pore plastics foam.

24. A process for producing a waste gas reactor according to any one of claims 12 to 22 comprising introducing a slurry of ceramic material into an open-pore plastics foam and then removing the plastics foam, wherein said non-noble metal oxide is added to a suspension, and the foam which is coated with ceramic material and which has been pre-cried is coated with the suspension, preferably by a dipping process, and then it is subjected to calcination.

25. A process according to claim 24 for producing a reactor according to claim 13 or any claim appendant thereto wherein the material (Al_2O_3) of the base layer is additionally mixed with the suspension.

26. A process for the production of a reactor according to any one of claims 12 to 22 comprising introducing a slurry of ceramic material into an open-pore plastics foam and then removing the plastics foam, wherein the foam which is coated with ceramic material and optionally a base layer and which is in a calcined or uncalcined condition is impregnated with a solution of a metal salt of a said non-noble metal and the metal salt is then converted into the metal oxide by chemical reaction and/or calcination, and fixed on the ceramic foam.

27. A process for the production of a reactor, according to any one of claims 23 to 26 and substantially as hereinbefore described.

28. An exhaust gas reactor substantially as herein described with reference to the accompanying drawings.

Printed for Her Majesty's Stationery Office by
Croydon Printing Company (UK) Ltd, 8-87, D8991685.
Published by The Patent Office, 25 Southampton Buildings, London, WC2A 1AY,
from which copies may be obtained.